

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re Application of

Yoshio Mitani, et al.

Serial No. 10/555,440

Art Unit: 1793

Filed: November 3, 2005

Examiner: Liano DIANA J

Title: HYDROPHOBIC FUMED SILICA

DECLARATION UNDER RULE 132

I, the undersigned Atsushi Takamuku, a citizen of Japan residing at c/o Tokuyama Corporation, 1-1 Mikage-cho, Shunan-shi, Yamaguchi-ken 745-8648, Japan, declare as follows:

I graduated from Hokkaido University in March, 1997, entered Tokuyama Corporation in April, the same year, and have engaged in the study and development of silica by the gaseous phase method since 1997 up to the present time.

I am one of the inventors of the instant patent application Serial No. 10/555,440, and am well versed in the contents of the invention disclosed and claimed in the instant application.

The Examiner in the Office Action cites Konya (US 2003/0103890) and denies the patentability of the instant invention under 35 USC 103. Namely, the hydrophobic fumed silica of the instant invention has the following properties:

- (a) An M-value is in a range of 48 to 65;
- (b) A tapping bulk density Td (g/L) is in a range of $80 < Td \leq 130$;
- (c) An n-value is in a range of 3.0 to 3.5 as measured in toluene; and
- (d) The content of aggregated particles having sizes of not smaller than 45 μm is not larger than 200 ppm.

On the other hand, Konya discloses a hydrophobic silica powder having an aerated bulk density in a range of 100 to 300 g, a primary particle size in a range of 10 to 120 nm and a hydrophobic degree in a range of 40 to 80 as measured by methanol titration. Besides, the hydrophobic silica powder of Konya is obtained by treating a hydrophilic silica powder obtained by a gaseous phase method with a cyclic siloxane to impart hydrophobic property thereto. Therefore, the Examiner takes a position that the hydrophobic fumed silica of the instant invention is simply specifying the properties which are essentially possessed by the hydrophobic silica powder of Konya, and denies the patentability of the instant invention.

However, as we have argued in our response to the Office Action of last time, the hydrophobic silica powder of Konya is obtained by mixing a hydrophilic silica powder obtained by a gaseous phase method with a liquid cyclic siloxane, followed by a compressing treatment by using a ball mill and, further, followed by treatment with an ammonia or an amine, which is a method quite different from the method of obtaining the hydrophobic fumed silica of the instant invention. Namely, the production method disclosed by Konya executes the compressing treatment after mixed with the cyclic siloxane, which is not the hydrophobic property-imparting treatment with the cyclic siloxane after the compressing treatment. Besides, Konya uses the cyclic siloxane in a liquid form instead of a gaseous form and, further, effects the compressing treatment relying on a mechanical milling by using a ball mill, which is not the compressing treatment free of shearing force that is attained by the compression by using a vacuum compressor. Therefore, the silica powder obtained by Konya does not have the above-mentioned properties (a) to (d) possessed by the hydrophobic fumed silica of the instant invention.

I, therefore, have conducted the following experiments

in order to prove the fact that the silica powder of Konya is different from the hydrophobic fumed silica of the instant invention.

<Preparation of the Starting Silica Powder>

As the silica powder for use in the following treatment, a hydrophilic silica powder A was prepared which was obtained by a gaseous phase method (flame method) and having the following properties. As the hydrophobic property-imparting agent, the following cyclic dimethylsiloxane was prepared.

Here, the hydrophilic silica powder having quite the same properties as the hydrophilic silica powder used in Examples of Konya was not available and, besides, the hydrophobic property-imparting agent, i.e., 1,3-dimethyl-1,3-bis(trimethylsiloxy)-1,3-dihydroxydisiloxane or hexamethylcyclotrisiloxane was not available. Therefore, there were prepared those having properties as close to the properties of them as possible.

Hydrophilic silica powder A:

BET specific surface area: 208 m²/g

Primary particle size: 15 nm (as measured by using a transmission electron microscope)

Aerated bulk density: 45 g/L

Hydrophobic property-imparting agent:

Octamethylcyclotetrasiloxane (cyclic dimethylsiloxane)

<Comparative Experiment 1>

By using the hydrophilic silica powder A, a hydrophobic fumed silica was produced in compliance with Example 1 of Konya.

That is, 0.5 kg of the hydrophilic silica powder A was fed into the Henschel's mixer of a content of 20 L, and 100 g of a mixed solution of 50 g of the hydrophobic property-imparting agent (octamethylcyclotetrasiloxane) and 50 g of

toluene was sprayed thereon with stirring at a rotational speed of 200 rpm to mix the hydrophobic property-imparting agent into the hydrophilic silica powder A.

The thus treated silica powder was fed into a 7-L ball mill containing alumina balls of a diameter of 20 mm, and was milled at a rotational speed of 50 rpm for 45 minutes to obtain a fine silica powder having an aerated bulk density of 130 g/L.

The thus obtained fine silica powder was fed into the Henschel's mixer of a content of 20 L, stirred and mixed at room temperature, heated while passing a mixed gas of nitrogen gas and ammonia (ammonia: 10% by volume) therethrough, maintained at a temperature of 200°C for 40 minutes, and was cooled to obtain a fine powdery hydrophobic fumed silica.

The powder was measured for its aerated bulk density, BET specific surface area and primary particle size and, at the same time, like in Example 1 of the instant application, was measured for its M-value, tapping bulk density Td, n-value in toluene and content of aggregated particles of sizes of not smaller than 45 μ m. The results were as shown in Table A.

<Comparative Experiment 2>

A fine powdery hydrophobic fumed silica was obtained by the same method as that of Comparative Experiment 1 but maintaining the heating temperature at 290°C while passing the mixed gas of nitrogen gas and ammonia.

The powder was measured for its properties in the same manner as in Comparative Example 1. The results were as shown in Table A.

That is, the octamethylcyclotetrasiloxane was used instead of 1,3-dimethyl-1,3-bis(trimethylsiloxane)-1,3-dihydroxydisiloxane described in Example 1 of Konya. In Comparative Example 1, therefore, the hydrophobic property could not be imparted to a sufficient degree. In

Comparative Example 2, therefore, the heating temperature while passing the mixed gas of nitrogen gas and ammonia was changed to 290°C.

<Comparative Experiment 3>

A fine hydrophobic fumed silica powder was obtained by the same method as that of Comparative Experiment 1 but changing the treating conditions by using the ball mill into a rotational speed of 50 rpm and a treating time of 20 minutes to obtain a fine silica powder having an aerated bulk density of 75 g/L and treating the fine silica powder with the mixed gas of nitrogen gas and ammonia.

The powder was measured for its properties in the same manner as in Comparative Example 1. The results were as shown in Table A.

That is, in Comparative Experiments 1 and 2, the tapping bulk densities were outside the range of the instant invention. Therefore, the conditions for compression treatment were varied to adjust the tapping bulk density so as to lie in the range of the instant invention, and the obtained hydrophobic fumed silica was evaluated for its properties.

Table A

		Comp. <u>Exp. 1</u>	Comp. <u>Exp. 2</u>	Comp. <u>Exp. 3</u>
BET specific surface area	m ² /g	179	168	168
Aerated bulk density	g/L	140	140	90
Primary particle size	nm	15	15	15
M-value		36	50	51
Tapping bulk density (Td)	g/L	145	145	95
n-Value		-	2.7	2.7
Aggregated particles of not smaller than 45 μm	ppm	1500	1300	1050

<Consideration>

As will be understood from the above experimental results, since the production method of Konya is different from the production method employed by the instant invention, the hydrophobic fumed silica obtained by Konya has properties different from those of the hydrophobic fumed silica of the instant invention. According to Konya, for example, the compressing treatment is conducted by the mechanical milling by using the ball mill and, therefore, the n-value and the content of aggregated particles of not smaller than 45 μm are lying outside the ranges of the instant invention. Further, even when the tapping bulk density is adjusted to lie in the range of the instant invention by varying the conditions of mechanical milling, the n-value and the content of aggregated particles of not smaller than 45 μm lie outside the ranges of the instant invention.

According to Konya as described above, the hydrophobic fumed silica of the instant invention is not obtained and, therefore, I believe that the patentability of the instant invention is not denied by Konya.

I, the undersigned Atsushi Takamuku, state that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment or both under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patents issuing thereon.

Date: *October 6, 2008*

Atsushi Takamuku

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